

Influence of orbital pair breaking on paramagnetically limited states in clean superconductors

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Abstract

Paramagnetic pair breaking is believed to be of increasing importance in many layered superconducting materials such as cuprates and organic compounds. Recently, strong evidence for a phase transition to the Fulde-Ferrell-Larkin-Ovchinnikov(FFLO) state has been obtained for the first time. We present a new theory of competing spin and orbital pair breaking in clean superconducting films or layers. As a general result, we find that the influence of orbital pair breaking on the paramagnetically limited phase boundary is rather strong, and its neglect seldom justified. This is particularly true for the FFLO state which can be destroyed by a very small orbital contribution. We discuss the situation in $\text{YBa}_2\text{Cu}_3\text{O}_7$ which has two coupled conducting Cu-O layers per unit cell. As a consequence, an intrinsic orbital pair breaking component might exist even for applied field exactly parallel to the layers.

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I. INTRODUCTION

Most theoretical studies of paramagnetic pair breaking in superconductors followed the attitude of the classical papers by Clogston¹ and Chandrasekhar² where only spin pair breaking was considered and the orbital component was assumed to be negligibly small. A notable exception is the dirty limit theory developed by Maki³, Fulde⁴ and others. In many experiments, on the other hand, *both* pair breaking components are present and the neglect of the orbital contribution is not really justified. Recently, ultra-thin films became available and several new classes of layered superconducting compounds have been discovered. For applied field parallel to the films⁵ or conducting planes⁶, Pauli paramagnetism can be the dominating pair breaking effect, provided the conducting layers are sufficiently separated from each other or the thickness of the films is sufficiently small. In many of these compounds, including High- T_c cuprates and organic superconductors, impurity scattering and spin-orbit coupling is small and orbital pair breaking is - for an applied field parallel to the planes - the most important second order effect, next to the spin effect, to be taken into account.

Of particular interest is the Fulde-Ferrell-Larkin-Ovchinnikov(FFLO) state^{7,8}, which is a spatially inhomogeneous superconducting state, predicted to occur in clean superconductors with purely paramagnetic limiting. Recent critical field measurements⁹ in the quasi-two-dimensional organic superconductor $\kappa - (\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ strongly suggest that a state of the FFLO type exists in this material; agreement between experiment⁹ and existing theories has been successfully checked¹⁰ both in view of the angle-dependence¹¹ and the temperature dependence¹² of the upper critical field (see also¹³). Apparently, this is the first time since the original predictions in 1964^{7,8} that quantitative agreement between theory and experiment with regard to the FFLO phase boundary has been established. Strong paramagnetic effects can also be expected for the High- T_c cuprate superconductors at low temperatures, when the conducting planes in adjacent unit cells are well separated from each other. A measurement^{6,14} at $T = 1.6\text{ K}$ in $\text{YBa}_2\text{Cu}_3\text{O}_7$ indicates rather clearly that the superconducting state is paramagnetically limited but, on the other hand, the observed

transition is too broad to allow a decision between the FFLO state and the homogeneous superconducting state.

A measure of the relative strength of orbital and paramagnetic pair breaking is the ratio of the paramagnetic critical field H_p divided by the orbital upper critical field H_{c2} of a type II superconductor. For a bulk superconductor in the clean limit this ratio can be written as

$$H_p/H_{c2} \sim \frac{\xi_0}{k_F^{-1}}, \quad (1)$$

in terms of the Fermi wavelength k_F and the coherence length ξ_0 of BCS theory.

This relation implies that orbital pair breaking will always be the dominating mechanism in bulk superconductors, no matter how large the Ginzburg-Landau (GL) parameter κ is; this holds at least in the framework of conventional BCS theory. In a thin superconducting layer of thickness $d < \xi_0$, on the other hand, the orbital critical field $H_c(d)$ is increased by a factor of ξ_0/d and the corresponding ratio is given by

$$H_p/H_c(d) \sim \frac{d}{k_F^{-1}}. \quad (2)$$

In comparison to Eq. (1) a small transverse dimension $d \ll \xi_0$ of the film suppresses the orbital effect and enlarges the spin effect drastically. However, equation (2) also shows, that the critical thickness which separates the spin pair-breaking and orbital pair-breaking dominated regimes is still of the order of an atomic distance. Thus, the estimate (2), which is confirmed by more quantitative calculations to be presented below, indicates that a nearly perfect two-dimensional situation is required in order to justify the neglect of orbital pair breaking contribution in clean superconductors. (The situation in dirty superconductors is much more favorable for the spin effect; the FFLO state, however, is suppressed by impurities).

The simultaneous action of both types of pair breaking has already been studied for a particular situation, an infinitely thin superconducting film in a tilted magnetic field^{15,11,16}. In such a configuration, orbital pair breaking is entirely due to the perpendicular field component, while the component parallel to the film is exclusively responsible for the spin

effect. The states near the upper critical field can be characterized by different Landau quantum numbers $n = 0, 1, 2, \dots$, depending on the tilt angle^{15,11}. The order parameter structure near H_{c2} has also been investigated and shows interesting properties, such as several zeros with different vorticity per unit cell¹⁶. For exactly plane-parallel external field the FFLO phase is recovered. However, in this limit no orbital pair breaking exists and only the spin effect survives, as a consequence of the vanishing thickness of the superconducting layer in this model.

In this paper we investigate a superconducting film of *finite thickness* in a magnetic field parallel to the conducting plane. Thus, the usual model of purely paramagnetic pair breaking is generalized in a different way, taking into account the influence of a finite orbital pair breaking component on the FFLO state. The model is formulated in section II, using the framework of the quasiclassical Eilenberger equations. We assume that the film thickness is smaller than the coherence length and use a cylindrical Fermi surface. This shape of the Fermi surface allows us to study the influence of orbital pair breaking without any additional complications, like scattering of quasiparticles at the film boundaries. Such boundary effects seem less important in the present context, but may, nevertheless, be present in many materials and should be taken into account in future work. In addition, the cylindrical shape of the Fermi surface, which corresponds to a truly two-dimensional situation, allows us to extend our investigations to superconducting layers of atomic dimension. The main results, obtained by solving numerically the relevant phase boundary and stability equations, and a discussion of possible orbital pair breaking contributions in the plane-parallel field configuration of $\text{YBa}_2\text{Cu}_3\text{O}_7$, are reported in section III. Finally, the results are summarized in section IV.

II. BASIC EQUATIONS

We first calculate the highest field where a superconducting solution of the quasiclassical equations, for small order parameter Δ , exists in a thin film. This field may correspond

to a second order phase transition or to the supercooling limit of the normal conducting state; to make a decision between these two possibilities the free energy of the competing homogeneous superconducting state will be calculated in a second step.

A. Stability limit of normal conducting state

Let the film be parallel to the xy plane with a finite extension from $-d/2$ to $+d/2$ in the z -direction. The applied magnetic field B is assumed to be parallel to the plane of the film and parallel to the y -direction, $\vec{B} = B\vec{e}_y$. The transport equations, linearized in Δ , are given by

$$\left[2(\omega_l - \imath\mu B) + \hbar\vec{v}_F(\hat{k}) \left(\vec{\nabla}_r - \imath(2e/\hbar c)\vec{A} \right) \right] f(\vec{r}, \hat{k}, \omega_s) = 2\Delta(\vec{r}, \hat{k}), \quad (3)$$

$$\left[2(\omega_l - \imath\mu B) - \hbar\vec{v}_F(\hat{k}) \left(\vec{\nabla}_r + \imath(2e/\hbar c)\vec{A} \right) \right] f^+(\vec{r}, \hat{k}, \omega_s) = 2\Delta^*(\vec{r}, \hat{k}), \quad (4)$$

Here, the Zeeman term μB occurs in the combination $\omega_s = \omega_l - \imath\mu B$, where $\omega_l = (2l+1)\pi k_B T$ are Matsubara frequencies, $\mu \simeq \hbar|e|/(2mc)$ is the magnetic moment of the electron and B is the magnitude of the induction. The self-consistency equation for the gap is given by

$$\left(2\pi k_B T \sum_{l=0}^{N_D} \frac{1}{\omega_l} + \ln \frac{T}{T_c} \right) \Delta(\vec{r}, \hat{k}) = \pi k_B T \sum_{l=0}^{N_D} \int d^2\hat{k}' V(\hat{k}, \hat{k}') \left[f(\vec{r}, \hat{k}', \omega_s) + f(\vec{r}, \hat{k}', \omega_s^*) \right], \quad (5)$$

where N_D is the cutoff index for the Matsubara sums. The Fermi velocity is given by $\vec{v}_F(\hat{k}) = v_F(\vec{e}_x \cos \varphi + \vec{e}_y \sin \varphi) = v_F \hat{k}$; the integral in Eq. (5) over the cylindrical Fermi surface is simply a one-dimensional integral over the angle variable φ . We allow for a separable gap anisotropy, $\Delta(\vec{r}, \hat{k}) = \Delta(\vec{r})\gamma(\hat{k})$, $V(\hat{k}, \hat{k}') = \gamma(\hat{k})\gamma(\hat{k}')$, which will be specialized later to s-wave and d-wave superconductivity. We use the following gauge for the vector potential: $A_x = Bz$, and $A_y = A_z = 0$.

The standard method to solve the linearized transport equations uses a complete set of eigenfunctions of the operator $\hat{k}\vec{\partial}_r$ to construct the inverse of the differential operators on the l.h.s. of Eqs. (3), (4). Here, $\vec{\partial}_r$ is an abbreviation for the gauge-invariant derivative,

$\vec{\partial}_r = \vec{\nabla}_r - i(2e/\hbar c)\vec{A}$. For a cylindrical Fermi surface, $\hat{k}\vec{\partial}_r$ contains no derivative with respect to z and the Green's functions depend on z in a purely local way (z playing the role of a parameter). This allows a straightforward generalization of the standard method to the present problem.

Let us start from the well-known bulk solution of eqs. (3),(4). If the eigenfunctions and eigenvalues of the operator $\hat{k}\vec{\partial}_r$ for an infinite sample are denoted by $f_{\hat{k},\vec{p}}$ and $i\hat{k}\vec{p}$ respectively,

$$\hat{k}\vec{\partial}_r f_{\hat{k},\vec{p}}(\vec{r}) = i\hat{k}\vec{p} f_{\hat{k},\vec{p}}(\vec{r}), \quad (6)$$

then the solution of Eq. (3) is given by

$$f(\vec{r}, \hat{k}, \omega_s) = \int d^3r_1 \int \frac{d^3p}{(2\pi)^3} \frac{f_{\hat{k},\vec{p}}^*(\vec{r}_1) f_{\hat{k},\vec{p}}(\vec{r})}{\omega_s + v_F \hat{k}\vec{p}/2} \Delta(\vec{r}_1, \hat{k}), \quad (7)$$

where the spatial integration extends over all space. In the chosen gauge the solutions of Eq. (6) are given by

$$f_{\hat{k},\vec{p}}(\vec{r}) = e^{-\frac{i}{2} \left[\frac{2|e|}{\hbar c} (\hat{k}\vec{r}) ((\vec{B} \times \vec{r})\hat{k}) + \kappa_{\parallel} z x \right] + i\vec{p}\vec{r}}, \quad (8)$$

with the abbreviation $\kappa_{\parallel} = \frac{2|e|}{\hbar c} B$. Using the completeness of the set of eigenfunctions (8) the Green's functions may immediately be written in the form of Eq. (7).

To transform relation (7) to a finite volume, it is, in our case, only necessary to restrict the spatial integration in Eq. (7) to the film volume, i.e. to perform the integration over z_1 from $-d/2$ to $+d/2$. This simple method works only for a cylindrical Fermi surface, where the momentum of the quasiparticles is always parallel to the film boundaries. Otherwise, quasiparticle scattering at the film boundaries leads, for small $d < \xi_0$, to a modification of the integral kernel which has to be calculated by solving Eq. (6) in a finite volume, with appropriate boundary conditions.

To proceed, the denominator of the integrand in Eq. (7) is shifted into an argument of an exponential function by means of the identity

$$\frac{1}{r} = \int_0^\infty dt e^{-rt}. \quad (9)$$

Now, if the eigenfunctions (8) are inserted, the Green's function (7) is represented as an integral with regard to the variables \vec{p} , \vec{r} , and t over an exponential function. Two of these integrations can be performed analytically and the Green's function takes the form

$$f(\vec{r}, \hat{k}, \omega_s) = \int_0^\infty dt e^{-t\omega_s} e^{-\frac{t}{2}v_F\kappa_{\parallel}z\hat{k}_x} \Delta(\vec{r} - \frac{t}{2}v_F\hat{k}, \hat{k}). \quad (10)$$

For $d < \xi$ the order parameter may be considered as z -independent and the Greens's function $f(\vec{r}, \hat{k}, \omega_s)$ may be replaced by its value $f(\hat{r}, \hat{k}, \omega_s)$ which depends only on x and y and denotes the average of $f(\vec{r}, \hat{k}, \omega_s)$, with respect to z , from $-d/2$ to $+d/2$:

$$f(\hat{r}, \hat{k}, \omega_s) = \int_0^\infty dt e^{-t\omega_s} \frac{1}{\frac{1}{4}tv_F\kappa_{\parallel}d\hat{k}_x} \sin\left(\frac{1}{4}tv_F\kappa_{\parallel}d\hat{k}_x\right) \Delta(\hat{r} - \frac{t}{2}v_F\hat{k}, \hat{k}). \quad (11)$$

The rest of the calculation is a straightforward generalization of methods developed in previous works^{8,11}. It is convenient to perform the following shift in the argument of the space-dependent part of the gap:

$$\Delta(\hat{r} - \frac{t}{2}v_F\hat{k}) = e^{-\frac{1}{2}tv_F\hat{k}\partial_{\hat{r}}} \Delta(\hat{r}). \quad (12)$$

Inserting the Green's function solutions in the self-consistency equation for the gap (5) and performing the Matsubara sum yields the linearized gap equation

$$\begin{aligned} -\ln\left(\frac{T}{T_c}\right) \Delta(\hat{r}) = \pi k_B T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int_0^{2\pi} \frac{d\varphi'}{2\pi} \gamma(\hat{k}')^2 \cdot \left[1 - \frac{1}{\frac{1}{4}tv_F\kappa_{\parallel}d\hat{k}'_x} \right. \\ \left. \cdot \sin\left(\frac{1}{4}tv_F\kappa_{\parallel}d\hat{k}'_x\right) \cos\left(\mu B t - \frac{1}{2t}tv_F\hat{k}'\hat{\nabla}\right) \right] \Delta(\hat{r}) \end{aligned} \quad (13)$$

which has to be solved in order to find the magnetic field where the normal-conducting state breaks down. The operator $\hat{\nabla}$ used in Eq. (13) acts in the x, y -plane. Eq. (13) differs from previous results¹¹ by a d -dependent factor which reduces to 1 in the limit $d \rightarrow 0$. For d-wave superconductivity the finite thickness of the film breaks rotational invariance; if Φ is the angle between the magnetic field and the y -axis of the crystal, the following replacement has to be performed in the integrand of Eq. (13):

$$d\hat{k}'_x \Rightarrow d\left(\hat{k}'_x \cos \Phi - \hat{k}'_y \sin \Phi\right), \quad (14)$$

in order to take the angle dependence of the external field into account.

To proceed further, the gap Δ is assumed to be proportional to plane wave states $e^{i\hat{q}\hat{r}}$. Solving the linearized gap equation (13), with $\hat{\nabla}$ replaced by $i\hat{q}$, for different wave numbers \hat{q} , one obtains a function $B(\hat{q})$. The field we are looking for, where the normal conducting state breaks down - and the corresponding wave number - is given by the highest $B(\hat{q})$.

Eq. (13) comprises two limiting cases where the behavior of the solutions is known, the purely paramagnetic limit (for $d = 0$), and the purely orbital limit (for $\mu = 0$). For $d = 0$ one obtains the standard FFLO result^{7,8}. For $\mu = 0$ Eq. (13) may be solved analytically near T_c . In this limit one obtains for the parallel critical field, where the normal-conducting solution breaks down

$$B_c^{\parallel} = 0.61 \frac{hc}{2e} \frac{\sqrt{1 - T/T_c}}{\xi_0 d}. \quad (15)$$

Eq. (15) is in agreement with the second order transition line obtained for $d < \xi(T)$ in the framework of GL theory. At low temperatures microscopic calculations have, to our knowledge, only been performed for Fermi surfaces of spherical¹⁷⁻¹⁹ or ellipsoidal¹⁹ shape. In these works a $d^{-3/2}$ behavior for the critical field has been obtained in the limit of very small d . In contrast, the present theory, using a cylindrical Fermi surface, leads to an approximate d^{-1} behavior of the critical field in the whole temperature range.

B. Phase boundaries of homogeneous states

For an infinitely thin film, the free energy of the FFLO state is only slightly lower than the free energy of the homogeneous (paramagnetically limited) superconducting state. The presence of an orbital pair breaking component, realized by a vector potential, in our film of thickness d may change the free energy balance in a decisive way. To clarify this point, we calculate the free energies of the homogeneous superconducting and normal-conducting states and compare the resulting phase boundaries and stability limits with the FFLO transition line studied in the last subsection. Considering films or layers of finite thickness, we

refer to states which do not depend on the coordinates x, y within the plane as ‘homogeneous states’; these states may, nevertheless, depend on z as a consequence of a (residual) screening property of the thin film.

The spatially constant, purely paramagnetically limited superconducting state has been studied first by Sarma²⁰; this case corresponds to $d = 0$ in the present model. For $T/T_c < 0.56$ he found a first order phase boundary, which lies below the FFLO transition line. This line is determined by inserting the solutions of the nonlinear gap equation

$$2\pi k_B T \sum_{l=0}^{N_D} \frac{1}{\omega_l} + \ln \frac{T}{T_c} = \pi k_B T \sum_{l=0}^{N_D} \left(\frac{1}{\sqrt{|\Delta|^2 + \omega_s^2}} + cc. \right), \quad (16)$$

into the free energy difference

$$F_s - F_n = -N(E_F) \pi k_B T \sum_{l=0}^{N_D} \left(\frac{(\omega_s - \sqrt{|\Delta|^2 + \omega_s^2})^2}{\sqrt{|\Delta|^2 + \omega_s^2}} + cc. \right). \quad (17)$$

For $T < 0.56 T_c$ the gap as a function of B has two branches^{20,21}, as shown in Fig. 1. Our calculation of the second variation of the free energy shows (see the curves for $T/T_c = 0.1$ in Fig. 1) that the homogeneous superconducting state may be superheated up to the highest field, where the two branches cross. The lowest field where the lower branch exists defines, on the other hand, the supercooling limit of the normal-conducting state. Thus, the region of the lower branch corresponds, as expected, exactly to the metastable region of the first order transition. In this way, three transition lines, the phase transition line where the free energies coincide, the superheating line, and the supercooling line, are determined by solving Eqs. (16), (17); for finite d the same method is used to determine the metastable region.

For the present circular Fermi surface, the determination of the ‘homogeneous states’ in a film of finite thickness d is still a local problem despite the nontrivial z -dependence appearing in the transport equations. The assumption of a gap which depends weakly on the z -coordinate leads, in analogy to the reasoning of the last subsection, to the following relation between the averaged Green’s functions \bar{f} , \bar{g} and the gap Δ :

$$\bar{f} = \frac{1}{B(d, \varphi)} \arctan\left(\frac{B(d, \varphi)}{\omega_s}\right) \Delta \bar{g}, \quad (18)$$

where

$$B(d, \varphi) = \frac{v_F k_{\parallel} d \cos \varphi}{4\pi k_B T_c}. \quad (19)$$

Note, that orbital pair-breaking leads to a dependence of the Green's functions on the quasiparticle wave number \hat{k} . Using Eq. (18) the self-consistency relation for the gap takes the form

$$2\pi k_B T \sum_{l=0}^{N_D} \frac{1}{\omega_l} + \ln \frac{T}{T_c} = \pi k_B T \sum_{l=0}^{N_D} \int_0^{2\pi} \frac{d\varphi}{2\pi} \left(\frac{\beta(\varphi)}{\sqrt{|\Delta|^2 \beta(\varphi) + A_l(d, \varphi)^2}} + cc. \right), \quad (20)$$

and the free energy difference is given by

$$F_s - F_n = -N(E_F) \pi k_B T \sum_{l=0}^{N_D} \int_0^{2\pi} \frac{d\varphi}{2\pi} \left(\frac{\left(A_l(d, \varphi) - \sqrt{|\Delta|^2 \beta(\varphi) + A_l(d, \varphi)^2} \right)^2}{\sqrt{|\Delta|^2 \beta(\varphi) + A_l(d, \varphi)^2}} + cc. \right), \quad (21)$$

where the factor $A_l(d, \varphi)$ is defined by

$$\frac{1}{A_l(d, \varphi)} = \frac{1}{B(d, \varphi)} \arctan\left(\frac{B(d, \varphi)}{\omega_s}\right). \quad (22)$$

The factor $\beta(\varphi)$ is 1 for s-wave and $1 + \cos(4\varphi)$ for d-wave superconductivity. Eqs. (20), (21) are essentially of the same (local) form as Eqs. (16), (17); the main difference is the dependence of the Green's functions on the direction φ of the quasiparticle momentum, which leads to the φ -integrals in Eqs. (20), (21). For $d \rightarrow 0$ the previous results are recovered as $A_l(d, \varphi)$ approaches ω_s in this limit. Solving Eq. (20) and calculating the free energy (21) the influence of a finite orbital pair breaking contribution, due to a nonzero film thickness d , on the three transition lines can be studied.

III. RESULTS AND DISCUSSION

In this section we discuss the following four transition lines, defined in more detail in the last section: (1) the line B_c where the free energies of the homogeneous superconducting and normal-conducting states coincide, (2) the superheating limit B_{sh} of the homogeneous superconducting state, (3) the supercooling limit (stability limit) of the normal-conducting state

against spatially homogeneous superconducting fluctuations, which is denoted by B_{sc} , and (4) the stability limit of the normal-conducting state against spatially inhomogeneous superconducting fluctuations, denoted by B_{FFLO} . The detailed results reported here are mainly for isotropic gap (s-wave) superconductors; a few calculations have also been performed for d-wave superconductors in view of recent experiments on $\text{YBa}_2\text{Cu}_3\text{O}_7$.

As a starting point, we show in Fig. 2 these four transition lines for $d = 0$, in the purely paramagnetic limit. For $T > T_{tri} = 0.56 T_c$ all four lines merge into a single second order transition line. Below the tricritical point T_{tri} only three different lines are visible in Fig. 2 since, interestingly, B_{FFLO} and B_{sh} exactly coincide (this coincidence occurs, however, only for a circular Fermi surface). Only B_{FFLO} is physically significant for $d = 0$, the other lines are meaningless. At B_{FFLO} a second order phase transition to the FFLO state takes place; the lower phase boundary of the FFLO state is not dealt with here; it has been studied by Burkhardt and Rainer²¹.

The paramagnetic pair-breaking effect dominates for very small d while the orbital effect dominates for large d . Thus, it should be possible to define a critical thickness d^* which roughly separates the two regimes. This crossover behavior is shown in Fig. 3 for the thermodynamic critical field B_c . The value of d^* is of the order of k_F^{-1} in the region of low T , in agreement with the estimate of section I. Fig. 3 shows also the decreasing importance of paramagnetic pair breaking with increasing T . Generally, the additional orbital pair-breaking effect brought about by the finite thickness of the conducting layer, leads to a depression of all four fields shown in the reference figure (Fig. 2). A detailed plot of the T -dependence of the fields B_{FFLO} , B_c , B_{sc} (the superheating field B_{sh} lies above B_{FFLO} and has been omitted for clarity) for $d/k_F^{-1} = 0.5, 1.0, 2.5$ as shown in Fig. 4 reveals, however, significant differences. The FFLO transition field is much stronger suppressed than the line B_c where the free energies of the homogeneous states coincide. As a consequence, for $d \gtrsim 0.5 k_F^{-1}$ (see top of Fig. 4) the FFLO transition vanishes (B_{FFLO} becomes the supercooling field of the normal state) and is replaced by a first order transition at B_c to the homogeneous superconducting state. Note that a conducting layer of atomic thickness

(dimension of unit cell in the plane) yields enough orbital pair breaking to produce this suppression of B_{FFLO} in favor of B_c . This behavior is not unreasonable; spatially varying states are known to be much more sensitive to perturbations than homogeneous states (recall in this context Anderson's theorem). With further increasing orbital pair breaking (see middle and bottom of Fig. 4) the lines B_{FFLO} and B_{sc} tend to merge and the metastable region shrinks; for $d > 3 k_F^{-1}$ orbital pair-breaking dominates.

The behavior of all four fields as a function of d is shown in Fig. 5 in the low temperature region (for $T = 0.01 T_c$), where paramagnetic effects are most pronounced. This figure gives an overview of the cross-over from the paramagnetically dominated regime at very small d to the orbitally dominated regime at large d . If the thickness where B_{FFLO} and B_c cross is denoted by d_1 , then the FFLO state is only realized in the small range $d < d_1 \cong 0.5 k_F^{-1}$, for $d > d_1$ a first order transition to the homogeneous (mainly) paramagnetically limited state occurs. The FFLO line plays the role of a supercooling limit of the normal state until it falls (at $d \cong 1.2 k_F^{-1}$) below the line B_{sc} , where the normal-conducting state is limited by spatially constant superconducting fluctuations. The wavenumber q of the FFLO state decreases with increasing d until it jumps to $q = 0$ at the line B_{sc} (at the crossing point two degenerate solutions exist for q). It should be pointed out that we continue to use here the term 'FFLO state', even if this term denotes, strictly speaking, a state without any orbital pair breaking contribution. With increasing d the lines B_{sh} , B_c , B_{sc} approach each other and the transition becomes identical to the well-known second order transition of a thin film in a parallel field, which is entirely due to orbital pair breaking (the region of really large d where the difference between type I and type II superconductivity becomes important is clearly outside the range of validity of the present model).

The results reported so far have been restricted to s-wave superconductors, with isotropic gap [using $\gamma(\hat{k}) = 1$ in Eqs. (13), (20)] . A few calculations have also been performed for d-wave superconductors, using $\gamma(\hat{k}) = 2(\hat{k}_x^2 - \hat{k}_y^2)$. The results generally confirm the behavior found for s-wave superconductors. An interesting peculiarity of d-wave superconductors without any orbital pair-breaking is a steep rise of B_{FFLO} with decreasing T below $T/T_c =$

0.1 (see Fig. 4 of Manalo and Klein¹⁰). This peak belongs to the $\phi_q = 0$ portion of the critical field¹² and is much steeper than the corresponding part of the critical field curve for s-wave superconductors. Our calculations show that this peak can be effectively suppressed by a very small ($d/k_F^{-1} \approx 0.2$) amount of orbital pair breaking. The absence of this peak in measurements⁹ on $\kappa - (\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ may be an indication of a very small residual orbital pair-breaking contribution in this material; a more detailed study would be worth while.

Let us investigate the consequences of the fact that the FFLO state can be suppressed in favor of the homogeneous superconducting state by a very small admixture of orbital pair-breaking (see Figure 5). We have not mapped out a complete phase diagram like figure 5 for d-wave superconductors (where B_{FFLO} becomes anisotropic as a consequence of the finite d) but instead performed a few calculations in order to get an overview of what happens. The results confirm qualitatively the main feature visible in figure 5, namely a much stronger suppression of B_{FFLO} , as compared to B_c , by orbital pair-breaking. For $\text{YBa}_2\text{Cu}_3\text{O}_7$ we have two conducting CuO_2 layers per unit cell with a distance of $\approx 3.9 \text{ \AA}$, while the c-axis zero-temperature coherence length is estimated²² to be $2 - 4 \text{ \AA}$. The coupling between bilayers in adjacent unit cells may obviously be neglected, as a consequence of the large length $c \approx 11.7 \text{ \AA}$ of the unit cell in this direction. The coupling between the two layers in one unit cell, on the other hand, remains an open question, and the following two possibilities should be taken into consideration.

The first possibility is, that the two superconducting layers decouple at low T , below some crossover temperature T^* . As is well known, the orbital critical field of weakly coupled layers diverges²³ below some crossover temperature T^* , which means that paramagnetic pair-breaking is the only remaining mechanism to limit the superconducting state. This requires a two-dimensional, in-plane mechanism of superconductivity. The amount of orbital pair-breaking would be negligibly small in this case and the superconducting state below the critical field should be the FFLO state.

The second possibility is that the superconducting state keeps its finite extension for

arbitrary T . This requires an inter-plane mechanism where the bilayer structure is essential for the superconducting pairing process. In this case, the bilayer may be approximately replaced by a single layer of finite thickness $d \approx 2 - 4 \text{ \AA}$. Taking a value of $v_F \approx 7 \cdot 10^7 \text{ cm/sec}$ for the Fermi velocity in the $a - b$ plane, as measured by Andreev reflections²⁴, we estimate a value between 1 and 4 for our dimensionless thickness parameter d/k_F^{-1} , which measures the amount of orbital pair-breaking. Thus in this case, if the bilayer structure can be approximated by a finite slab, the amount of intrinsic orbital pair-breaking in YBCO, brought about by the finite thickness of this slab, will be by far large enough to suppress the FFLO state. The second order FFLO transition will be replaced by a first order transition to a homogeneous superconducting state; this transition is due to the combined action of *both* pair breaking mechanisms rather than a single one. For a typical value of $d/k_F^{-1} \approx 2$, the critical field would be still of the same order of magnitude as the purely paramagnetically limited, (Pauli limiting) field at $d = 0$ but with a strongly reduced metastability region (see Fig. 5)

The transport measurement of Dzurak et al.⁶ of the critical field of $\text{YBa}_2\text{Cu}_3\text{O}_7$ at 1.6K led to a result of the order of the Pauli limiting field $B_p = B_c(d = 0)$; the transition seems, however, too broad to distinguish between the FFLO and Pauli limiting fields. Further, more accurate experiments are required to settle this question, which concerns fundamental aspects of the superconducting state in High- T_c cuprates. Observation of the FFLO state in the plane-parallel field configuration of $\text{YBa}_2\text{Cu}_3\text{O}_7$ would be a strong argument in favor of an in-plane mechanism of superconductivity in this material. The relevance of this question has also been discussed by Yang and Sondhi²⁵, using the framework of the Lawrence-Doniach model, which is in a sense complementary to the present approach.

The orbital pair-breaking effect due to the finite thickness of the conducting layers has previously been taken into account in a theory by Schneider and Schmidt²⁶. This theory may be used successfully to fit the experimental data¹⁴ near T_c , but neglects all paramagnetic effects. The purely paramagnetic limit, on the other hand, has been studied by Maki and Won¹² and by Yang and Sondhi²⁷. The present results show that *both* effects should be

taken into account for a detailed description of the transition. The orbital effect cannot be neglected even if the orbital critical field is several times higher than the paramagnetic limiting field.

Finally, we note, that single atomic (molecular) layers are responsible for the superconducting state in the organic compound $\kappa-(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ where phase boundaries compatible with a d-wave version of the FFLO state have recently been observed. Thus, a considerable influence of orbital pair-breaking, which would suppress the FFLO state in favor of the homogeneous superconducting state, does not exist in this material. Measurements of the detailed temperature-dependence of B_{FFLO} close to $T = 0$ could reveal even small admixtures of an orbital pair-breaking component.

IV. CONCLUSION

We developed a theory of competing paramagnetic and orbital pair-breaking effects in clean superconducting films and layers. The destructive influence of orbital pair-breaking on the FFLO state turned out to be stronger than commonly expected. It is necessary to have single-atomic layers in exactly plane-parallel fields in order to be able to neglect completely the orbital component and observe the “pure” FFLO state. We calculated only the upper phase boundary of the “mixed” FFLO state (which occurs in films of finite thickness as a consequence of the combined action of both pair breaking mechanisms). The equilibrium structure as well as the lower transition line to the homogeneous superconducting state, have not yet been calculated for this new inhomogeneous state. Another challenging open question is a microscopic treatment of Josephson coupling between layers of finite thickness. Our results show that a careful measurement of the plane-parallel critical field in $\text{YBa}_2\text{Cu}_3\text{O}_7$ could give important information on the question whether an in-plane or an inter-plane mechanism is responsible for superconductivity in High- T_c superconductors.

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FIGURES

FIG. 1. Stable and unstable branch of Δ as a function of B for $t = T/T_c = 0.1$ and second derivative of free energy $F_{\Delta^*\Delta}$ for $t = 0.1, 0.3, 0.6$.

FIG. 2. Critical fields B_{FFLO} , B_c , and B_{sc} as a function of T for $d = 0$. The superheating field B_{sh} agrees with B_{FFLO} for the present circular Fermi surface.

FIG. 3. Critical field B_c as a function of the dimensionless thickness parameter k_F^{-1}/d for $t = T/T_c = 0.5$ and $t = 0.9$. The purely orbital pairbreaking curve, which is proportional to d^{-1} and the purely paramagnetic curve, which is independent of d are shown as dotted lines ($t = 0.5$).

FIG. 4. Critical fields B_{FFLO} , B_c , B_{sc} as a function of T for $d/k_F^{-1} = 0.5$ (top), $d/k_F^{-1} = 1.0$ (middle), and $d/k_F^{-1} = 2.5$ (bottom).

FIG. 5. Critical fields B_{FFLO} , B_c , B_{sc} , B_{sh} as a function of d for $T = 0.01 T_c$.













